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13. ABSTRACT (Maximum 200 words) The goal of our research program is to elucidate the hot electron physics of alternating-current thin-film electroluminescent (ACTFEL) devices with particular emphasis on: 1) hot electron transport, 2) defect state hot electron sourcing and trapping, and 3) hot electron-induced ACTFEL aging. Highlights of our research accomplishments are summarized in terms of these three topical categories.			
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**HOT ELECTRON PHYSICS OF ALTERNATING-CURRENT THIN-FILM
ELECTROLUMINESCENT DEVICES: PHASE II**

Final Report

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HOT ELECTRON PHYSICS OF ALTERNATING-CURRENT THIN-FILM ELECTROLUMINESCENT DEVICES: PHASE II

I. Statement of the Problem Studied

The goal of our research program is to elucidate the hot electron physics of alternating-current thin-film electroluminescent (ACTFEL) devices with particular emphasis on: 1) hot electron transport, 2) defect state hot electron sourcing and trapping, and 3) hot electron-induced ACTFEL aging. Highlights of our research accomplishments are summarized in terms of these three topical categories.

II. Summary of Important Results

1. Hot Electron Transport

(a) Monte Carlo Simulation of High-Field Transport in ZnS

Monte Carlo simulation of high field transport in ZnS is performed. Several major refinements have been incorporated into the present version of the Monte Carlo simulation program, as discussed in the following.

In previous simulations, a nonparabolic, three-valley model¹ or a local empirical pseudopotential method (EPM) calculation² was employed for the conduction band. Currently, the conduction band density of states is calculated using the nonlocal EPM method in which the calculated reflectivity spectrum is fit to recent UV spectroscopic ellipsometric data.³ The gamma valley effective mass is calculated to be $0.17 m_0$, in reasonable agreement with a recent cyclotron resonance measurement⁴ of $0.20 \pm 0.01 m_0$. Note that both the nonlocal EPM and cyclotron resonance effective masses are significantly different than the 0.39 and 0.28 estimates that arise from work performed in the 1960's. The nonlocal EPM calculation yields estimates of the gamma-to-L and gamma-to-X intervalley separation of 1.27 and 0.83 eV, respectively, which differ significantly from the 1.48 and 1.48 eV estimates obtained from a local EPM calculation.

Previous conduction band models have included only the 1st and 2nd conduction bands, up to 5.3 eV above the conduction band minimum. The 3rd and 4th bands, as obtained from a nonlocal EPM calculation of the band structure, are included in the present model up to approximately 12 eV above the conduction band minimum. The resulting density of states shows two distinct jumps at ~ 0.9 and ~ 1.4 eV, corresponding to the gamma-to-L and gamma-to-X intervalley separation energies. Also, a peak and a hump in the density of states exist at ~ 2.4 and ~ 4 eV, respectively.

In previous simulations, band-to-band impact ionization was either ignored¹ or was modeled using a simple Keldysh formulation,² given by: $R_{ii}(E) = R(E_{th}) [(E-E_{th})/E_{th}]^2$ where $R(E_{th})$ is the total phonon scattering rate at the threshold energy, E_{th} . In the present model, an empirical fit to a numerical, energy-dependent, full-band impact ionization rate computed directly from a quantum-kinetic Boltzmann equation by Professor Redmer of Rostock University using our nonlocal EPM band structure is used.^{5,6} The deduced empirical fit equation is given by: $R_{ii}(E) = 5.1944e10 (E-E_{th})^{5.075}$ where $E_{th} = 3.8$ eV.

Inclusion of the four conduction bands and revision of the formulation of the band-to-band impact ionization rate results in a hot electron tail which extends out to ~6-7 eV and a small hump at ~4 eV in the hot electron distribution which corresponds to the ~4 eV hump in the conduction band density of states. The hot electron tail occurs at a higher energy for the present modeling than in previous models; this is primarily the result of a smaller band-to-band impact ionization rate, which does not reach a rate of 1e15 /sec until energies in excess of ~8 eV. (In contrast, when the Keldysh model is used, a scattering rate of ~1e15 /sec occurs at an energy of ~4.5 eV. It is clear from this and from previous calculations that the calculated hot electron distribution is extremely sensitive to how band-to-band impact ionization is modeled since this is the primary mechanism for cooling of the hot electron distribution when the electrons reach high energies.)

For completeness, spin-orbit splitting is included in the Monte Carlo model. It appears that spin-orbit splitting is not very important in determining the high field transport properties of ZnS. This is not surprising since the spin-orbit splitting is very small (0.064 for ZnS) and the fields of interest for TFEL operation are so large.

Currently, the greatest source of uncertainty in the Monte Carlo simulation of high-field transport in ZnS is related to the deformation potentials, which are the model parameters accounting for the strength of the electron-phonon interaction. The deformation potential values used in the present version of the Monte Carlo simulation are obtained from a comparison of the calculated ionization coefficient with different choices of the deformation potential to experimental carrier multiplication data.

One of the unique features of the present version of the Monte Carlo model is the inclusion of impact excitation of Mn²⁺ using a simplified version of the theoretical treatment of exchange scattering, as derived by Bringuer and Bhattacharyya.⁷ The simplification used involves assuming Gaussian orbitals for the ground and excited states of the impurity center so that a closed form analytical expression for the impact excitation scattering rate is obtained for use in the simulation. Using this formulation, the simulated scattering rates for impact excitation and impact ionization are plotted as a function of phosphor field. The simulation shows that the threshold for both impact excitation and impact ionization occur at about 1 MV/cm, in good agreement with the electroluminescent threshold found experimentally from photo-induced luminescence (PIL) measurements discussed in Section II.1.d.

(b) Monte Carlo Simulation of High-Field Transport in GaN and SrS

A Monte Carlo study of high-field transport in GaN is undertaken⁸ in order to assess the importance of the electronic bandstructure in determining high-field transport properties. GaN is chosen because it is the best studied wide bandgap semiconductor known, in terms of bandstructure, with a bandgap comparable to ZnS. In this comparative study, a nonlocal empirical pseudopotential bandstructure and impact ionization rates are calculated for GaN, and then a GaN Monte Carlo simulation is performed using the same deformation potential parameters as used for ZnS in order to determine the role of the bandstructure. The Monte Carlo simulation predicts GaN to be a better transport material than ZnS, yielding a more energetic hot electron distribution. The critical high-field transport issue clarified by this study is simply that a lower conduction band density of states leads to a more energetic hot electron distribution (assuming identical deformation potentials). A peak in the conduction band density of states is found to lead to a corresponding pile-up in the hot electron distribution at a similar energy.

Concisely, this study, and our collective high-field transport work to date, suggests that the intrinsic high-field transport properties (i.e. properties not related to electron injection or to impact ionization of deep level traps) of relevance for ACTFEL applications depend primarily on three properties of the phosphor: (i) the conduction band density of states; a smaller density of states leads to a more energetic hot electron distribution; (ii) the band-to-band impact ionization rate; the smaller the impact ionization rate and the larger its energy threshold, the more energetic the hot electron distribution since band-to-band impact ionization is the primary physical mechanism which cools the electron distribution under the very high-field conditions of relevance for ACTFEL operation; and (iii) the magnitude of the deformation potential; the larger the deformation potential (i.e. the strength of the electron-phonon interaction), the less energetic the hot electron distribution.

A preliminary study of the intrinsic high-field transport properties (i.e. properties not related to electron injection or to impact ionization of deep level traps) of SrS has been initiated. To date, two SrS Monte Carlo simulations have been performed, based on two different assumptions about the SrS bandstructure. The first simulation is performed assuming atomic-like pseudopotential parameters whereas the second simulation is performed using nonlocal pseudopotential parameters fit to an APW calculation of the SrS bandstructure.¹⁰ In both cases, the impact ionization rates are obtained from the calculated bandstructure and the deformation potentials are assumed to be identical to that of ZnS, for comparative purposes since nothing is currently known about the deformation potentials of SrS.

Radically different simulation results are obtained, depending on which bandstructure is employed in the Monte Carlo simulation. The bandstructure obtained assuming atomic-like pseudopotential parameters, which results in conduction bands that are relatively flat (i.e. atomic-like), leads to the conclusion that SrS is a very poor transport material and that phosphor fields of ~3-4 MV/cm are required before a significant number of hot electrons are available for impact excitation or impact ionization of luminescent impurities. In contrast, the bandstructure based on fitting of the pseudopotential parameters to the calculated APW bandstructure leads to a hot electron distribution that is more energetic than ZnS, suggesting that SrS is an excellent transport

material for ACTFEL applications. Thus, at this time, it is not possible to obtain a reliable Monte Carlo high-field transport simulation of SrS because of uncertainties in the bandstructure, and also due to uncertainties in the deformation potential. We attempted to address these bandstructure uncertainties experimentally by measuring the reflectivity spectra of SrS (see Section II.2.d); however, the measured reflectivity spectra are dominated by exciton absorption, so that very little relevant information about the details of the bandstructure could be deduced.

Three lines of investigation are currently being pursued so that accurate SrS Monte Carlo high-field transport simulations may be performed in the future. First, photo-induced luminescence (PIL) measurements (see Section II.1.d) of SrS ACTFEL devices are in progress to determine the threshold field at which impact excitation of Cu and impact excitation or ionization of Ce luminescent impurities is initiated; this information will serve as empirical calibration data for the Monte Carlo simulation. Second, first principles, ab initio calculations of the SrS bandstructure are being performed, by Professor Vogl and co-workers at the Technical University in Munich using a local density approximation approach. This approach promises to yield a more reliable SrS bandstructure; additionally, this theoretical approach promises the possibility of being able to calculate the effects of excitons in the reflectivity spectra so that experimental reflectivity spectra can be more reliably be interpreted in terms of bandstructure information. Third, first principles calculations of the deformation potential are in progress; these calculations offer the promise of being able to unambiguously deduce the deformation potential directly, given the bandstructure of the phosphor .

(c) Impact Excitation Quantum Yield

Recently we devised a new measurement technique, denoted hot electron impact excitation quantum yield, for the assessment of the impact excitation quantum yield as a function of the phosphor field.¹¹ This experiment relies on the use of a field-control circuit to control the magnitude of the phosphor field. This technique has been employed for the assessment of green ZnS:Tb, yellow ZnS:Mn, and blue thiogallate:Ce ACTFEL devices.

A comparison of the impact excitation quantum yield of ZnS:Tb ACTFEL devices grown by atomic layer epitaxy (ALE) and by sputtering reveals that the ALE devices exhibit a strong temperature-dependence whereas the temperature-dependence of the sputtered device is very weak. It is suspected that the high degree of temperature sensitivity of the ALE device is related to the temperature dependence of the radiative recombination efficiency of the excited luminescent impurity. Additionally, the impact excitation quantum yield possesses a threshold of ~0.5 MV/cm and a saturation of ~1.5 MV/cm.

Impact excitation quantum yield experiments have been performed on ZnS:Mn and thiogallate:Ce samples. The ZnS:Mn devices exhibit a threshold at ~0.7 MV/cm and some evidence for the onset of saturation at ~2-2.5 MV/cm. The thiogallate:Ce samples exhibit a threshold at ~1.0 MV/cm and very strong saturation at ~1.7 MV/cm at low temperature, but no evidence for saturation at room temperature. The thresholds for both of these samples appear to be temperature-independent; this suggests that these thresholds arise from tunnel emission and are not transport-determined.

These impact excitation quantum yield experiments were devised for calibrating the Monte Carlo high-field transport simulations so that some of the physical parameters which are least well established (notably the deformation potentials, which are parameters accounting for the strength of intervalley scattering) may be appropriately adjusted to obtain agreement between measured data and simulation. Some comparisons between simulated and experimental data have been made.^{3,12} Currently, we are no longer pursuing the impact excitation quantum yield experiment because it is difficult to interpret whether the measured thresholds are determined by the transport or charge-injection properties of the ACTFEL device. We now perform the photo-induced transferred charge (PIQ) and photo-induced luminescence (PIL) measurements for this kind Monte Carlo calibration purpose (see following section).

(d) Photo-Induced Transferred Charge and Photo-Induced Luminescence

Photo-induced-charge (PIQ) and photo-induced-luminescence (PIL) measurements are employed for the characterization of evaporated ZnS:Mn ACTFEL devices.¹³ PIQ and PIL measurements involve monitoring the charge and luminescence associated with the transport of UV-excited electrons or holes under the application of a constant applied voltage. The experiment is accomplished using a short UV laser pulse to create carriers near the upper phosphor/insulator interface while maintaining a constant voltage across the ACTFEL device in order to set the phosphor field. By changing the polarity of the applied voltage pulse, either electron or hole transport may be studied. The PIQ/PIL measurement was pioneered by Corlatan et al.^{14,15}

PIQ and PIL experiments are performed using evaporated ZnS:Mn ACTFEL devices with three different phosphor thicknesses; 300, 700, and 950 nm. The PIL thresholds indicate that the critical field for initiating Mn impact excitation in ZnS is ~1.04 MV/cm for electron-initiated impact excitation. The existence of a PIL threshold under a condition in which hole transport dominates suggests that Mn impact excitation can be hole-initiated. However, we have determined that this hole-transport-dominated PIL threshold actually arises from electron injection and transport of electrons emitted from the interface opposite from where UV absorption occurs. Trapped holes give rise to band bending which increases the cathode field at the opposite interface, facilitating interface electron injection. This conclusion is established by demonstrating that the hole-transport-dominated PIL threshold increases with decreasing UV intensity, which is attributed to a decrease in hole trapping and a concomitant increase in the threshold for interface electron injection.

The PIQ trends indicate that electron transport is significantly more efficient than hole transport. This trend is interpreted as evidence for hole trapping in the ZnS, consistent with the conclusions of Corlatan et al.^{14,15} If it is assumed that a negligible amount of electron trapping occurs, a comparison of electron and hole PIQ curves, an average drift distance for holes prior to being trapped of ~200 nm may be deduced. Furthermore, this drift distance can be used to deduce an average hole lifetime of ~2 ps. Finally, the hole trap capture cross section can be estimated to be $\sim 7 \times 10^{-14} \text{ cm}^2$. This is a relatively large capture cross section, corresponding to a negatively charged deep level whose charge state leads to very efficient hole capture due to the attractive

coulombic interaction between the trap and the hole. It is speculated that this hole trap is most likely a defect complex involving a zinc vacancy.

2. Defect State Hot Electron Sourcing and Trapping

(a) Electrical Characterization

(i) Yellow ZnS:Mn ACTFEL Devices

A comparison of the operation and electrical characteristics of ZnS:Mn ACTFEL devices grown by evaporation and atomic layer epitaxy (ALE) is accomplished.¹⁶ Evaporated devices have symmetric electrical characteristics with respect to the applied voltage polarity and exhibit strong field-clamping. In contrast, ALE devices are asymmetric, exhibit space charge creation during operation, and do not show strong field-clamping. The asymmetry and space charge characteristics found in the ALE ACTFEL devices are attributed to the presence of a nonuniform distribution of zinc vacancies in the ZnS phosphor near to the top interface and also to asymmetries in the interface state density of the two interfaces.

C-V, Q-F_p, and Q-V analysis of three ALE ZnS:Mn ACTFEL devices with different phosphor thicknesses (thin, normal, and thick) is performed as a function of temperature (100, 200, 300, 400 K).^{17,18} The devices are pre-aged 24 hours to stabilize their characteristics. C-V and Q-F_p analysis indicates that all samples show evidence for space charge generation. The least amount of space charge generation occurs at the lowest temperature and the amount of space charge generation increases with increasing temperature. The thicker the sample, the larger the amount of space charge generation and the stronger the temperature-dependence of the space charge generation. The existent data suggests that a process involving phonon-assisted tunneling is determining the temperature-dependence of these devices (Note: A process involving band-to-band or deep level impact ionization should have an opposite temperature-dependence to that measured for these samples and a process involving pure tunneling should be temperature-independent.)

The operation of ACTFEL devices may be strongly affected by the presence of dynamic or static positive space charge within the phosphor layer during device operation.¹⁹ Dynamic space charge is positive charge in the phosphor layer which is periodically created and annihilated during each period of the applied voltage waveform. In contrast, static space charge is positive space charge in the phosphor layer whose charge state does not change appreciably during steady-state operation of the ACTFEL device. The static space charge density of evaporated ZnS:Mn ACTFEL devices is estimated to be $\sim 7 \times 10^{16} \text{ cm}^{-3}$ from measured trends in the phosphor clamping field as a function of phosphor thickness. This static space charge density estimate implies a cathode clamping field of $\sim 2.2 \text{ MV/cm}$ and a clamping interface trap depth of $\sim 1.5 \text{ eV}$. Furthermore, from transferred charge trends as a function of the phosphor thickness it is concluded that static space charge in evaporated ZnS:Mn ACTFEL devices arises from metastable hole trapping in the phosphor. It is speculated that the physical origin of these metastable hole traps are zinc vacancies or zinc vacancy complexes.

A new method for accomplishing transferred charge analysis of ACTFEL devices is proposed.²⁰ The measurement is accomplished using an applied voltage waveform in which a large number of bipolar voltage pulses with a constant maximum applied voltage (V_{max}) above threshold (these pulses are referred to as setting pulses) are inserted between measuring pulses of variable V_{max} . The transferred charge, Q_{max} , is monitored only during the measuring pulse. The purpose of the setting pulses is to ensure that the initial polarization charge state is constant for each measurement of Q_{max} . Because the setting pulse measurements are performed with a constant initial polarization charge, they exhibit transferred charge characteristics closer to ideal than conventional transferred charge measurements. The utility of the setting pulse transferred charge technique is demonstrated via the analysis of evaporated ZnS:Mn and atomic layer epitaxy SrS:Ce ACTFEL devices. A setting pulse transferred charge derivative curve (or transferred charge C-V curve) exhibits significantly less overshoot than does a conventional transferred charge derivative curve since less space charge creation occurs during the measuring pulse when setting pulses are employed. A comparison of setting pulse and conventional transferred charge curves provides useful information regarding the build-up of space charge in the phosphor layer. The transferred charge characteristics of evaporated ZnS:Mn and ALE SrS ACTFEL devices appear to arise from the build-up of static and of dynamic space charge, respectively.

(ii) Blue Thiogallate:Ce ACTFEL Devices

$Q-F_p$, C-V, and $Q_{max}-V_{max}$ measurements are performed over a temperature range of 150-540 K.²¹ This work was motivated, in part, by the observance of thermal quenching of blue thiogallate:Ce ACTFEL panels as the panel temperature rose due to the close positioning of the electronic drivers. As the temperature increases, the conduction charge increases, the threshold decreases, the lower portion of the C-V transition shifts rigidly, and the apparent insulator capacitance (as measured from C-V curves) increases to values in excess to that of the physical insulator capacitance (at room temperature, the apparent insulator capacitance is less than that of the physical insulator capacitance). From $Q-F_p$ trends, it appears that either the insulator or phosphor capacitance changes as a function of temperature (it is suspected that the phosphor capacitance is temperature-dependent). However, the observed changes, if attributed to the phosphor capacitance, are of an inadequate magnitude to account for thermal quenching. The thermal quenching may be associated with: #1 a decrease in the luminescent impurity excitation efficiency with increasing temperature, or #2 a decrease in the radiative recombination efficiency of the luminescent impurities with increasing temperature.

(iii) Blue SrS:Ce ACTFEL Devices

Sputtered SrS:Ce ACTFEL devices with three different thicknesses (400 , 700, 1250 nm) are assessed via Q-V, C-V, and $Q-F_p$ analysis. No overshoot (space charge generation in a phosphor is typically manifest as overshoot in C-V and $Q-F_p$ plots) is observed for the two thinner samples; a very small amount of overshoot is evident for the thickest sample. This result is very different from what we observe for ALE SrS:Ce ACTFEL devices which always exhibit extremely large amounts of overshoot.²² The steady-state phosphor field, as estimated from $Q-F_p$ analysis, is reduced with increasing phosphor thickness; this is evidence that space charge generation is indeed present

in these sputtered SrS:Ce ACTFEL devices. In summary, there appears to be some space charge generation present in sputtered SrS:Ce ACTFEL devices, but the amount space charge generation is significantly less than for ALE devices; also, there seem to be some differences in how these two different kinds of devices manifest space charge generation. The space charge in sputtered SrS is believed to be more static than dynamic in nature.

(b) Space Charge Modeling

A discrete sheet of charge model, which accounts for the presence of space charge generation in the phosphor layer of certain ACTFEL displays, is developed.²³⁻²⁵ Two different mechanisms for the creation of bulk space charge, involving field-emission and impact ionization, are implemented in the model. The model is capable of simulating overshoot in C-V or Q-F_p characteristics of ACTFEL devices with moderate amounts of overshoot [e.g. yellow ZnS:Mn ACTFEL devices grown by ALE]. Various overshoot trends, such as the extent of overshoot as a function of the position of the sheet and the density of traps in the sheet, have been examined.

Sheet charge modeling is helping to clarify subtle issues regarding ACTFEL internal device operation and is leading to new ways of modeling ACTFEL behavior. Two new SPICE equivalent circuit models for ACTFEL devices which possess space charge generation are proposed. These models are deduced by mapping the sheet charge device physics models to corresponding equivalent circuits. Two models are required to account for the two physical processes, field-emission and impact ionization, responsible for the generation of space charge. In the SPICE equivalent circuits, interface state injection and bulk charge injection are both represented by nonlinear, voltage-controlled current sources (VCCSs). Impact ionization charge generation is represented by a series combination of a VCCS and an amplifier with a nonlinear gain, corresponding physically to charge multiplication by impact ionization.

(c) State-Space Modeling

The primary motivation for the development of a state-space model²⁶ for ACTFEL devices is to perform a realistic simulation of the steady-state electrical characteristics of evaporated ZnS:Mn ACTFEL devices as deduced from Q-V or Q-F_p measurements. We have found it to be a very challenging task to simulate the Q-V and Q-F_p characteristics as a function of the maximum applied voltage (V_{max}) of evaporated ZnS:Mn ACTFEL devices driven by a bipolar applied voltage waveform.

Initial simulations, which employed a discrete interface state, could not adequately account for the turn-on voltage, leakage charge, or relaxation charge trends of measured Q-F_p curves. Next, efforts were made to account for these deficiencies through the inclusion of a distributed interface state density. It was thought that an appropriate distributed interface state density could at least account for leakage charge (i.e. the charge which flows between voltage pulses) since it seemed evident that leakage charge must arise from electron emission of more shallow interface states as the phosphor field is significantly reduced during the interpulse interval. Indeed, various assumed interface state distributions did give rise to variable amounts of leakage charge. However, the

unavoidable conclusion yielded by these simulations is that no realistic, or even unrealistic distribution of interface states could provide simulations that would simultaneously account for experimental leakage charge and turn-on voltage trends.

In order to obtain realistic Q-V and Q-F_p simulations, it is necessary to include holes into the simulation. In the simulation it is assumed that holes are created via band-to-band impact ionization, are instantaneously captured in bulk trap states at a single energy level within the bandgap, are eventually re-emitted from bulk traps via phonon-assisted tunneling, and then drift to the cathode where they are annihilated via recombination with interface state electrons. In order to keep the model as simple as possible, it is assumed that space charge due to the hole occupancy of bulk trap states is uniform across the phosphor and that the total space charge density depends on the hole capture and re-emission history.

The inclusion of holes has allowed a realistic simulation of Q-V and Q-F_p curves to be accomplished. One surprising result is that the simulation implies that leakage charge arises from the re-emission of holes from bulk traps and their subsequent recombination with interface state electrons; usually leakage charge is attributed to electron emission from shallow interface states. Moreover, the re-emission of holes accounts for turn-on voltage trends; the turn-on voltage increases with hole re-emission and annihilation at the interface since this reduces the internal polarization charge. Additionally, holes account for experimental relaxation charge trends; relaxation charge arises from hole recombination with interface state electrons during the V_{max} portion of the waveform.

(d) Reflectivity Measurements

Reflectivity measurements of ZnS, SrS, and thiogallate TFEL phosphors are undertaken at the Army Research Laboratory (ARL), Fort Monmouth, New Jersey, using a vacuum ultraviolet (VUV) spectrophotometer in order to determine bandstructure information of the thiogallate phosphor.²⁷ Three different sulfides are measured and three distinctly different kinds of reflectivity spectra are obtained:

I: ZnS: The reflectivity spectrum is typical of group IV, III-V, and II-VI semiconductors, which are taken to be "covalent" materials, even though the ionicity of ZnS is 0.623. The spectrum is characterized as slowly rising just above the bandgap with two well-defined peaks occurring at ~2 and ~3.3 eV above the bandgap.

II: SrS: The reflectivity spectrum is typical of ionic materials such as alkali halides and titanates. Most of the "oscillator strength" occurs in two very strong exciton peaks located ~0.25 and 0.7 eV above the bandgap; the intensity of the reflectivity spectrum then drops precipitously up to about 3.5 eV above the bandgap.

III: Thiogallates: The reflectivity spectrum rises monotonically and is structureless up to ~4 eV above the bandgap where there is a broad peak. The best explanation, to date, for the thiogallate spectra's lack of structure is that these materials possess positional disorder even though they are

stoichiometric. All of the thiogallates measured to date have very similar reflectivity spectra, even when they are compositionally dissimilar.

These reflectivity results suggest that the bandstructure of these sulfides, and hence, presumably their transport properties, are very different. Thus, it does not appear likely that differences in the transport properties of these sulfides alone can explain why sulfides are the best ACTFEL phosphors to date. It is believed that it is the peculiar charge sourcing properties of these sulfides, associated with the presence of static or dynamic space charge, which makes them so appropriate for ACTFEL applications.

(e) Excitation / De-excitation Measurements

Excitation/de-excitation spectra of doped and undoped SrS phosphors grown by different methods is performed at ARL in order to investigate the nature of the defects or the luminescent impurities present in these materials. A detailed comparative study of SrS:Ce phosphors grown by multi-source deposition (MSD) (from Heinrich-Hertz Institut, Berlin, Germany) and ALE (from Planar International) is undertaken. A very strong infrared peak at 700 nm (1.77 eV) is observed for the emission spectrum of the doped and the undoped ALE sample but is not present for the MSD sample. This infrared peak is believed to be related to a larger concentration of native defects in the SrS grown by ALE compared to that grown by MSD.

3. Hot Electron-Induced ACTFEL Aging

(a) Yellow ALE ZnS:Mn ACTFEL Devices

Aging studies of ALE ZnS:Mn ACTFEL devices with different phosphor thicknesses (thin, normal, and thick) are performed by acquiring C-V, Q-F_p, and Q-V data as a function of aging time.¹⁷ The aging characteristics of the normal thickness sample are virtually identical to what we have reported previously.²⁸ Aging of the normal thickness device is found to be partially due to changes near the phosphor/insulator interfaces but also to strongly depend on changes in the bulk ZnS phosphor; these aging trends are denoted as interface- and bulk-related. Note that aging of the normal thickness sample is primarily bulk-related (preliminary results of an aging study of the thin phosphor device indicate that aging of this device is primarily bulk-related also). The aging characteristics of the normal thickness device are ascribed to an enhanced presence of Cl near the Al electrode phosphor/insulator interface and its tendency to migrate into the ZnS during aging. This type of aging is sometimes referred to as n-shift, since the brightness-voltage (B-V) curve tends to shift in a negative direction towards lower threshold voltages.

The nature of the aging is found to be distinctly different for the thick phosphor sample. In contrast to the normal and thin samples, which exhibit bulk-related aging characteristics, the thick sample possesses aging characteristics which are primarily interface-related (e.g. C-V curves which rigidly shift to higher voltages, a decrease of polarization charge with aging). This result is very surprising since thick devices are found to possess much more space charge generation than normal thickness samples; thus, more bulk charge sourcing would be expected for the thicker

devices. Apparently, even though the thicker phosphor device does exhibit more space charge generation, this bulk space charge generation does not exhibit large changes with aging. The experimental aging trends found in the thicker device are very similar to what we have reported previously for evaporated ZnS:Mn TFEL devices.²⁹ Aging of these evaporated devices, and also of the thick ALE device investigated in this study, is attributed to the formation of deep level, fixed charge states at the phosphor/insulator interface which are created as a consequence of interfacial atomic interdiffusion. The creation of these deep level, fixed charge states leads to a reduction of both conduction and polarization charge with aging. One attractive feature of this type of aging (which is sometimes referred to as p-shift, since the B-V threshold shifts to more positive voltages with aging) is that it saturates so that the threshold voltage eventually does not change appreciably with aging time. In contrast, n-shift aging does not exhibit saturation.

Another aging trend involves offset of Q-V or Q-F_p curves. According to the single sheet charge model,²³⁻²⁵ offset of a Q-V or Q-F_p curve from the origin is evidence that the centroid of space charge generation is located away from the center of the phosphor. The normal thickness device exhibits an appreciable offset prior to aging and for short aging times but the amount of offset is significantly reduced as aging progresses. This is interpreted as indicating that for the normal thickness device, the charge generation centroid is initially offset toward the top phosphor/insulator interface and the charge generation centroid moves toward the center of the phosphor as aging progresses. This movement of the charge centroid is believed to be associated with distribution of Cl with aging. In contrast, the thick device exhibits almost no Q-V or Q-F_p offset, suggesting that the charge generation centroid is very close to the center of the phosphor layer; the more symmetrical nature of the electrical characteristics and aging trends with respect to applied voltage polarity support this hypothesis. Since Cl would undoubtedly be present in the phosphor layer of the thicker device, this suggests that the Cl distribution is more uniform in the phosphor of the thicker device.

(b) Blue Thiogallate:Ce ACTFEL Devices

Q_{max} -V_{max} and Q-F_p measurements are performed²¹ as a function of aging time at a frequency of 1 kHz for a maximum aging time of 16 hours. Q_{max} -V_{max} aging trends are identical to B-V aging trends. This result is important for two reasons. First, it establishes that the aging characteristics for these devices are determined by electronic processes, not optical processes. Second, Q_{max} -V_{max} measurements are easier to automate than B-V measurements. The threshold voltage shifts rigidly to higher voltages with aging time (i.e., a so-called p-shift) and saturates after about 5 hours (i.e. most of the aging occurs in the first 5 hours). The conduction charge decreases and the phosphor field increases with aging time. Also, the shape of the Q-F_p curve near the threshold changes with aging in a manner that is consistent with charge injection occurring from deeper interface states as aging progresses. All of these aging trends are very similar to trends reported in an aging study of evaporated ZnS:Mn ACTFEL devices.¹⁷ Therefore, tentatively the same aging mechanism is invoked for blue thiogallate:Ce ACTFEL devices as proposed previously for yellow, evaporated ZnS:Mn ACTFEL devices; namely, hot electron recombination at the insulator/phosphor interface leading to atomic rearrangement / interdiffusion at the phosphor /insulator interface with a concomitant creation of deep level, fixed charge states. Note that it is very desirable that thiogallate devices exhibit a p-shift and interdiffusion/fixed charge aging mechanism since the most stable ACTFEL

devices manufactured to date possess this kind of an aging characteristic.

III. Conclusions and Directions of Future Work

The goal of our research program is to elucidate the hot electron physics of ACTFEL devices with particular emphasis on: 1) hot electron transport, 2) defect state hot electron sourcing and trapping, and 3) hot electron-induced ACTFEL aging. A significant amount of progress has been made in all three of these areas of research emphasis, as described in this Final Report.

Future experimental and theoretical research is planned with the goal of elucidating the hot electron physics of ACTFEL devices in order to further enhance the development of a full-color flat-panel display technology. Three areas are identified in which the primary research activity will be focused: hot carrier (electrons and holes) transport, hot carrier sourcing and trapping, and hot electron-induced aging. Whereas the first two phases of this research program concentrated on ZnS:Mn (yellow), thiogallate:Ce (blue), and SrS:Ce (blue-green) ACTFEL devices, the central thrust of the third phase of this research program will be to explore the hot carrier physics of ACTFEL devices employing a new SrS:Cu blue phosphor and a two-layer stack white phosphor (ZnS/SrS). Additionally, future research effort will be extended to include investigation of organic light-emitting devices (OLEDs). Future work will be performed in collaboration with Planar Systems, Inc. and the Army Research Laboratory.

IV. List of Publications

1. J. F. Wager, W. M. Ang, S. Pennathur, L. Pham, S. M. Goodnick, and A. A. Douglas, "Impact Ionization in Evaporated ZnS:Mn Alternating-Current Thin-Film Electroluminescent Devices", Proceedings of the International Symposium on Inorganic and Organic Luminescence, October 5-6, 1994, Hamamatsu, Japan.
2. J.F. Wager, "A Comparison of ZnS:Mn ACTFEL Devices Prepared by Evaporation and Atomic Layer Epitaxy", Proceedings of the 7th International Workshop on Electroluminescence (EL'94), October 10-12, 1994, Beijing China.
3. P. D. Keir, W. M. Ang, and J. F. Wager, "Modeling Space Charge in Alternating-Current Thin-Film Electroluminescent Devices using a Single Sheet Charge Model," *J. Appl. Phys.* 78, 4668-4680 (1995).
4. P. D. Keir, W. M. Ang, and J. F. Wager, "Modeling Space Charge in ACTFEL Devices using a Single- Sheet-Charge Model," *SID 95 Digest*, pg. 476-479 (1995).
5. R. L. Thuemler, P. D. Keir, and J. F. Wager, "Phosphor Field Dependence in ALE SrS:Ce ACTFEL Devices," *SID 95 Digest*, pg. 473-475 (1995).
6. P. Rack, P. Holloway, S. S. Sun, E. Dickey, C. King, L. Pham, and J. F. Wager, "Brighter Blue Electroluminescent Devices with an Enhanced Electron Injection Layer," *SID 95 Digest*,

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7. W. M. Ang, S. Pennathur, L. Pham, J. F. Wager, S. M. Goodnick, and A. A. Douglas, "Evidence for Band-to-Band Impact Ionization in Evaporated ZnS:Mn Alternating-Current Thin-Film Electroluminescent Devices," *J. Appl. Phys.* 77, 2719 (1995).
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10. J. A. Samuels, D. C. Smith, K. N. Siebein, K. Salazar, R. T. Tuenge, C. F. Shaus, C. N. King, H. Le, J. Hitt, R. L. Thuemler, and J. F. Wager, "MOCVD of SrS and SrS:Ce Thin Films for Electroluminescent Flat Panel Displays," Materials Research Society Meeting, (1995).
11. I. Lee, S. Pennathur, K. Streicher, T. K. Plant, J. F. Wager, P. Vogl, and S. M. Goodnick, "High-Field Electron Transport of the ZnS Phosphor in AC Thin-Film Electroluminescent Devices," *Inst. Phys. Conf. Ser. No. 145*, 1229 (1996).
12. S. M. Goodnick, M. Dur, and S. Pennathur, "Monte Carlo Simulations of High Field Transport in Inorganic EL", in Inorganic and Organic Electroluminescence, edited by R. H. Mauch and H.-E. Gumlich (Wissenschaft und Technik Verlag, Berlin), p. 8 (1996).
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Eckstein, and W. Schattke, "Impact Ionization Rate and High Field Transport in ZnS with Nonlocal Band Structure," *J. Appl. Phys.* **80**, 5054 (1996).

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19. R. Myers and J. F. Wager, "Transferred Charge Analysis of Evaporated ZnS:Mn Alternating-Current Thin-Film Electroluminescent Devices," *J. Appl. Phys.* **81**, 506 (1997).

20. T.K. Plant, U. Troppenz, B. Huettl, K.O. Velthaus, and R.H. Mauch, "Electro-Optic Excitation/De-Excitation Effects in SrS:Ce ACTFEL Devices from 15K - 300K," *J. SID* **4**, 287 (1996).

21. U. Troppenz, T.K. Plant, B. Huettl, K.O. Velthaus, and R.H. Mauch, "Electrical Properties of SrS:Ce EL Devices," *J. SID* **4**, 293 (1996).

22. J. F. Wager and P. D. Keir, "Electrical Characterization of Thin-Film Electroluminescent Devices," a chapter in *Annual Review of Materials Science* **27**, 228(1997).

23. J. C. Hitt, J. F. Wager, and S. S. Sun, "Static Space Charge in Evaporated ZnS:Mn Alternating-Current Thin-Film Electroluminescent Devices," *J. Appl. Phys.* (submitted).

24. J. C. Hitt, J. Peery, P. D. Keir, J. F. Wager, S. S. Sun, S. Moehnke, and R. T. Tuenge, "A Novel Method for Performing Transferred Charge Analysis of Alternating-Current Thin-Film Electroluminescent Devices," *J. Appl. Phys.* (submitted).

25. B. A. Cleary, J. C. Hitt, P. D. Keir, T. K. Plant, J. F. Wager, and S. S. Sun, "Photo-Induced Charge and Luminescence Measurements of Evaporated ZnS:Mn Alternating-Current Thin-Film Electroluminescent Devices," *J. SID* (submitted).

26. M. Dur, S. M Goodnick, S. S. Pennathur, J. F. Wager, M. Reigrotzki, and R. Redmer, "High-Field Transport and Electroluminescence in ZnS Phosphor Layers," *J. Appl. Phys.* (submitted).

V. List of Technical Presentations

1. Plenary lecturer: J. F. Wager, "Thin-Film Electroluminescence: A Tutorial", Advanced Displays Technologies International School, August 28-September 4, 1994, Lviv, Ukraine.

2. Invited talk and panelist: J. F. Wager, "Impact Ionization in Evaporated ZnS:Mn Alternating-Current Thin-Film Electroluminescent Devices", International Symposium on Inorganic and Organic Luminescence, October 5-6, 1994, Hamamatsu, Japan.

3. Invited talk: J. F. Wager, "Alternating-Current Thin-Film Electroluminescence: An Overview," Korea Institute of Science and Technology, October 7, 1994, Seoul, Korea.
4. Invited talk: J. F. Wager, "Impact Ionization and a Comparison of ZnS:Mn ACTFEL Devices," Gold Star Central Research Laboratory, October 8, 1994, Seoul, Korea.
5. Invited talk: J.F. Wager, "A Comparison of ZnS:Mn ACTFEL Devices Prepared by Evaporation and Atomic Layer Epitaxy", 7th International Workshop on Electroluminescence (EL'94), October 10-12, 1994, Beijing China.
6. Poster paper: P. D. Keir, W. M. Ang, and J. F. Wager, "Modeling Space Charge in ACTFEL Devices using a Single Sheet Charge Model," Society for Information Display International Symposium, Orlando, Florida, May 23-25, 1995.
7. Poster paper: R. L. Thuemler, P. D. Keir, and J. F. Wager, "Phosphor Field Dependence in ALE SrS:Ce ACTFEL Devices," Society for Information Display International Symposium, Orlando, Florida, May 23-25, 1995.
8. Poster paper: P. Rack, P. Holloway. S. S. Sun, E. Dickey, C. King, L. Pham, and J. F. Wager, "Brighter Blue Electroluminescent Devices with an Enhanced Electron Injection Layer," *Society for Information Display International Symposium*, Orlando, Florida, May 22-26, 1995.
9. Contributed talk: I. Lee, S. S. Pennathur, S. M. Goodnick, and J. F. Wager, "Band Structure and High-Field Electronic Transport in Alternating-Current Thin-Film Electroluminescent Devices," Spring Meeting of the Materials Research Society, San Francisco, April 17-21, 1995.
10. Seminar: S. M. Goodnick, "High Field Transport in ZnS Phosphors," Marburg, Germany, June 26, 1995 and Osaka University, Japan, September 18, 1995.
11. Contributed Talk: S. Pennathur, I. Lee, K. Streicher, T. K. Plant, J.F. Wager, P. Vogl, and S.M. Goodnick, "Hot Electron Excitation of Luminescent Impurities in AC Thin-Film Electroluminescent (ACTFEL) Devices," 9th International Conference on Hot Carriers in Semiconductors, Chicago, Illinois, July 31-August 4, 1995.
12. Invited Talk: S. M. Goodnick, S. Pennathur, I. Lee, and J. F. Wager, "High-Field Transport of the ZnS Phosphor in AC Thin-Film Electroluminescent Devices," 22nd International Symposium on Compound Semiconductors, Cheju Island, Korea, August 28-September 2, 1995.
13. Contributed talk: M. Peter, K. Lite, K. Streicher, T. K. Plant, and J. F. Wager, "Hot Electron Impact Ionization in Thin-Film Electroluminescent Devices," First International Conference on the Science and Technology of Display Phosphors, San Diego, California, November 14-16, 1995.
14. Contributed Talk: T. K. Plant, U. Troppenz, B. Hutt, K. O. Velthaus, and R. H. Mauch, "Electro-Optic Excitation/De-Excitation Effects in SrS:Ce ACTFEL Devices from 15 to 300K,"

1st International Conference on the Science and Technology of Display Phosphors, San Diego, California, November 14-16, 1995.

15. Contributed Talk: J. A. Samuels, D. C. Smith, K. N. Siebein, K. Salazar, R. T. Tuenge, C. F. Shaus, C. N. King, H. Le, J. Hitt, R. L. Thuemler, and J. F. Wager, "MOCVD of SrS and SrS:Ce Thin Films for Electroluminescent Flat Panel Displays," Materials Research Society Meeting, Boston (1995).
16. Invited talk: J. F. Wager, P. D. Keir, R. T. Thuemler, and S. Shih, "Thin-Film Electroluminescent Device Space Charge Modeling," First International Conference on the Science and Technology of Display Phosphors, San Diego, California, November 14-16, 1995.
17. Invited talk: J. F. Wager, "Characterization and Modeling of Thin-Film Electroluminescent Devices," American Ceramic Society's 1996 Annual Meeting and Exposition, Indianapolis, IN, April 14-17, 1996.
18. Contributed talk: J. F. Wager, "Electroluminescent Phosphors: Point Defects", Eighth International Workshop on Electroluminescence, Berlin, Germany August 13-15, 1996.
19. Invited talk: S. M. Goodnick, M. Dur, and S. Pennathur, "Monte Carlo Simulations of High Field Transport in Inorganic EL", Eighth International Workshop on Electroluminescence, Berlin, Germany August 13-15, 1996.
20. Contributed talk: B. Huttl, P. Kratzert, K. Lite, B. Reinserger, T. K. Plant, and R. H. Mauch, "Study of Transport in SrS:Ce, Mn, Cl Electroluminescent Devices by Optical Method," Eighth International Workshop on Electroluminescence, Berlin, Germany August 13-15, 1996.
21. Contributed talk: J. Hitt and J. F. Wager, "Static Space Charge in Evaporated ZnS:Mn TFEL Devices," Second International Conference on the Science and Technology of Display Phosphors, San Diego, California, November 18-20, 1996.
22. J. Peery, P. D. Keir, W. M. Ang, and J. F. Wager, "Simulation of Evaporated ZnS:Mn ACTFEL Devices," Third International Conference on the Science and Technology of Display Phosphors, Huntington, Beach, California, November 3-5, 1997.
23. B. A. Cleary, P. D. Keir, J. C. Hitt, T. K. Plant, J. F. Wager, and S. S. Sun, "Photo-induced Charge and Luminescence in Evaporated ZnS:Mn ACTFEL Devices," Third International Conference on the Science and Technology of Display Phosphors, Huntington, Beach, California, November 3-5, 1997.
24. M. Dur, S. M. Goodnick, M. Reigrotzki, and R. Redmer, "A Comparative Study of High-Field Transport in ZnS- and SrS-Based TFEL Devices," Third International Conference on the Science and Technology of Display Phosphors, Huntington, Beach, CA, November 3-5, 1997.

VI. List of Participating Scientific Personnel

1. Manoj Kumar
 - (i) M.S. Thesis: "Electrical Characterization and Aging Studies of Green ZnS:Tb AC Thin-Film Electroluminescent Devices"
 - (ii) Currently employed at: Exar Corporation, San Jose, California
2. K. Streicher
 - (i) M.S. Thesis: "Electro-Optic Characterization of ZnS:Tb³⁺ ACTFEL Devices for Probing the Hot Electron Distribution"
 - (ii) Currently employed at: Exar Corporation, San Jose, California
3. L. Pham
 - (i) M.S. Thesis: "Electrical Characterization, Maximum Charge-Maximum Voltage (Qmax-Vmax) Analysis, Aging, and Temperature Studies of Thin-film Electroluminescent Thiogallate Devices"
 - (ii) Currently employed at: Intel Corporation, Aloha, Oregon
4. S. S. Pennathur
 - (i) Ph.D. Thesis: "Monte Carlo Device Modeling Applications on Parallel Computers"
 - (ii) Currently employed at: Altera Corporation, San Jose, California
5. S. Shih
 - (i) M.S. Thesis: "Electrical Characterization and Aging Studies of ALE ZnS:Mn ACTFEL Devices with Varying Phosphor Layer Thicknesses"
 - (ii) Currently employed at: LSI Logic, San Jose, California
6. W. M. Ang
 - (i) Ph.D. Thesis: "Novel ACTFEL Phosphors Deposited by RF Sputtering" (in progress)
 - (ii) Currently employed at: Sharp Microelectronics Technology, Inc., Camas, Washington
7. P. D. Keir
 - (i) M.S. Thesis: "Modeling Phosphor Space Charge in Alternating-Current Thin-Film Electroluminescent Devices"
 - (ii) Currently a Ph.D. Candidate, Oregon State University
* Awarded Outstanding Research Assistant, OSU College of Engineering, 1996

* Intel Fellow, 1997

8. Insook Lee
 - (i) Postdoc
 - (ii) Currently employed at: LG Electronics Research Center, Seoul, Korea
9. Manuela Peter
 - (i) M.S. Thesis: "Impact Excitation Efficiency in AC-Driven Thin-Film Electroluminescent Devices"
 - (ii) Currently a Ph. D. Candidate at Tottori University, Tottori, Japan
10. Kevin Lite
 - (i) M.S. Thesis: "Optical Characterization of Alternating-Current Thin-Film Electroluminescent Phosphors and Devices"
 - (ii) Currently employed at Wacker-Siltronics, Portland, Oregon
11. Manfred Dur
 - (i) Postdoc (OSU/ASU)
12. Robert Thuemler
 - (i) M. S. Thesis: "Characterization of Alternating-Current Thin-Film SrS:Ce Electroluminescent Devices"
 - (ii) Currently employed at: Intel Corporation, Aloha, Oregon
13. John C. Hitt
 - (i) M. S. Thesis: "Static Space Charge in Evaporated ZnS:Mn Alternating-Current Thin-Film Electroluminescent Devices"
 - (ii) Currently a Ph.D. Candidate, Oregon State University
14. Hieu Le
 - (i) M. S. Thesis: (in final stages of preparation)
 - (ii) Currently employed at: Intel Corporation, Aloha, Oregon

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14. D. Corlatan, K. Neyts, P. De Visschere, and J. Van den Bossche, in *Proceedings of 1994 International Workshop on Electroluminescence*, edited by X. Xu (Science Press, Beijing, 1994),

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